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Prediction of disclinations in nematic elastomers

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The isochoric extension of a cylindrical, nematic-elastomeric specimen is studied. Numerical solutions of the resulting boundary-value problem predict that, for sufficiently large extensions, there exists a region surrounding the cylinder axis wherein molecular asphericity vanishes. This region is bounded by a narrow transition layer across which the asphericity drops rapidly and attains a non-trivial negative value. It is also found that there is a marked drop in the energy density across the transition layer. A state of this type corresponds to a disclination of strength +1 located along the cylinder axis. The disclination core is associated with the region of vanishing asphericity, in which the polymeric molecules are shaped as spherical coils, and the core radius is taken to be the location of the energy drop. At a generic point outside of the core radius, the polymeric molecules are shaped as ellipsoidal coils of revolution oblate about the cylinder radius. For realistic values of the material parameters, the energy criterion yields a core of approximately 0.015 μm, which is consistent with observations in conventional liquid-crystal melts.

1 Introduction

As technological applications of liquid crystals have grown over the past thirty years, so too has research into the structure and importance of their defects. Initially, defect-induced disruptions were the bane of liquid crystal displays. Now though, as Mottram and Sluckin [1] observe, these very defects are being harnessed in the zenithal bistable display. Also, as shown by Chandrasekhar [2] and Kléman [3], the presence of defects is necessary for the stabilization of some of the so-called blue phases observed in cholesteric liquid crystals. In addition, recent research by Mottram and Sluckin [1], Adrienko and Allen [4], and Mottram and Hogan [5] has shown the important effect of disclinations on the phase transition behavior in nematic liquid crystals. Since point, line, and surface defects manifest themselves in liquid crystals and, as noted by Kléman [3], their study has had enhanced the understanding of defects in other media.

A disclination in a nematic liquid crystal is a line along which the director is undefined. In the Oseen–Zöcher–Frank (OZF) theory (Oseen [6], Zocher [7], Frank [8]), a non-integrable singularity in the free-energy density occurs at a disclination. Historically, this difficulty was addressed by positing a core of fixed radius and energy about the disclination. Both Mottram and Hogan [5] and Ericksen [9] rightly conclude though that the fixed energy approach gives no information about the magnitude and energy of the core and also fails to elucidate the underlying physical nature of the core. Nevertheless, for nematics confined to capillaries, it has been shown by Williams et al. [10] that the director can 'escape into the third dimension' at the singularity, thus obviating the need for a core but not necessarily ruling one out altogether. This deficiency of the OZF theory led Ericksen [9] to develop a regularized theory involving the degree-of-orientation, a scalar field which vanishes at disclinations and enters the free energy density in a manner that mollifies the singularity otherwise associated with a disclination. Exploiting this idea, Mottram and Sluckin [1], Adrienko and Allen [4], and Mottram and Hogan [5] have obtained values of the core radius and energy of a disclination in a cylindrical configuration.

Here, we study disclinations in nematic elastomers. These materials differ from melts in that the polymer chains in the elastomer are cross-linked. The nematic mesogens may still be either main-chain or pendant, as in a melt. Nematic and other optically active polymers are of ever growing interest, as is attested by the reviews of Finkelmann [11], Zentel [12], Davis [13], Warner & Terentjev [14], and Terentjev [15], and the recent work on artificial muscle by deGennes et al. [16] and on tunable lasers by Finkelmann et al. [17]. Our effort is motivated by the belief that the understanding of disclinations and other defects in optically active polymers such as nematic elastomers is of fundamental importance and will have far-reaching technological consequences.

2 Model

We envision the elastomer to be formed in a two-step process. First, the nematic melt is cross-linked in a uniaxial state, i.e., one in which the average molecular conformation of the polymeric molecules at each material point assumes the shape of an ellipsoid of revolution. An annealing process is then performed. This process randomizes the orientation of nematic mesogens and results in an isotropic state—taken to be our reference state—wherein the conformation at each material point is spherical.

To describe such a material, we consider a simplification and simultaneous extension of the molecular-statiscal free-energy density of Warner, Gelling and Vilgis [18]. The simplification stems from restricting conformations to the uniaxial case and taking the reference state to be isotropic. On the other hand, the extension follows from including a term analogous to that arising in the OZF theory and terms associated with the degree of asphericity of the ellipsoids of revolution. These include a double-well potential in the asphericity and a regularizing term quadratic in the gradient of the asphericity. The double-well potential isolates as preferred not only the isotropic reference state but also those states with asphericity equivalent to that present at the time of cross-linking. In this sense, the material "remembers" the asphericity present at the instant of cross-linking. Specifically, the free-energy density has the form

$$\frac{\mu}{2}\Big((1+q)^{\frac{1}{3}}\Big(|\boldsymbol{F}|^2-\frac{q}{1+q}|\boldsymbol{F}^{\top}\boldsymbol{n}|^2\Big)-3\Big) + \frac{\kappa q^2}{2(1+q)^2}|\boldsymbol{F}^{\top}\boldsymbol{G}|^2+f(q)+\frac{\alpha}{2}|\boldsymbol{h}|^2, \qquad (1)$$

where: $\boldsymbol{F}$ is the deformation gradient, subject to the constraint $\det\boldsymbol{F}=1$ of incompressibility; $\boldsymbol{n}$ is the nematic director of unit magnitude; $\boldsymbol{G}=\operatorname{Grad}\boldsymbol{n}$ is the director gradient; $q>-1$ is the asphericity (the case of $-1<q<0$ corresponds to average molecular conformation having the shape of an oblate ellipsoid of revolution about $\boldsymbol{n}$, while prolate ellipsoids of revolution about $\boldsymbol{n}$ are described by $q>0$); and $\boldsymbol{h}=\operatorname{Grad}q$ is the asphericity gradient. In addition, $\mu>0$ is the rubber elasticity modulus, $\kappa>0$ the nematic elasticity modulus, and $\alpha>0$ a regularizing modulus. Finally, f is a double-well potential which we take to have the specific form

$$f(q)=\frac{\nu q^2(q-q_*)^2}{2(1+q)^2}, \qquad (2)$$

with $\nu>0$ determining the height of the energy barrier between states with $q=0$ and $q=q_*$. When $q=0$, the material is isotropic; otherwise, it is anisotropic. The case of $q=q_*$ corresponds to the asphericity present at the time of initial cross-linking. The rightmost term of (1) regularizes the theory and, granted the presence of f, allows for the existence of states in which q varies smoothly between its energetically preferred values: 0 and q_*.

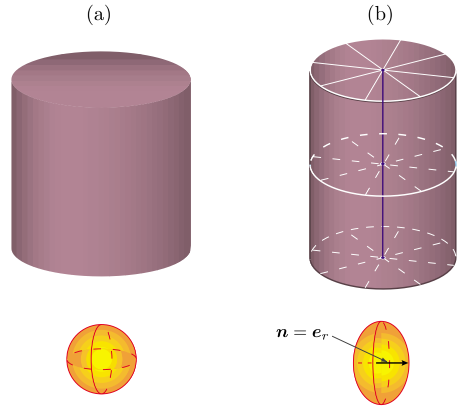

Figure 1: Cylinder and molecular conformation in undistorted (a) and distorted (b) states.

We use (1) to investigate the presence of disclinations of strength +1 in a cylindrical, nematic-elastomeric specimen of radius R and length L subjected to an isochoric deformation with gradient

$$\boldsymbol{F}=A\boldsymbol{e}_r\otimes\boldsymbol{e}_r+A\boldsymbol{e}_\theta\otimes\boldsymbol{e}_\theta+\frac{1}{A^2}\boldsymbol{e}_z\otimes\boldsymbol{e}_z, \qquad (3)$$

where $A>0$ is the ratio of the deformed to undeformed cylinder radius. Hence, values of A between zero and unity correspond to extension of the cylinder along its axis while those greater than unity correspond to contraction. We suppose that the director is either radial, viz.,

$$\boldsymbol{n}=\boldsymbol{e}_r, \qquad (4)$$

or, as would be the case when $q=0$, undefined. Further, we suppose that the asphericity depends at most on the radial coordinate r.

Introducing $x=r/R$ and $Q(x)=q(Rx)$ and using our assumptions concerning $\boldsymbol{F}$, $\boldsymbol{n}$, and q, we arrive at the dimensionless specialization

$$\frac{\mu}{2\nu}\Big((1+Q)^{\frac{1}{3}}\Big(A^2\Big(\frac{2+Q}{1+Q}\Big)+\frac{1}{A^4}\Big)-3\Big) + \frac{\kappa A^2Q^2}{2R^2\nu x^2(1+Q)^2}+\frac{Q^2(Q-q_*)^2}{2(1+Q)^2}+\frac{\alpha}{2\nu R^2}\Big(\frac{dQ}{dx}\Big)^2. \qquad (5)$$

of the free-energy density. The terms in (5) correspond, in order, to *rubber elasticity* ψ_e, *nematic elas-*

ticity ψ_n, *memory* ψ_m, and *regularization* ψ_r, and we write $\psi = \psi_e + \psi_n + \psi_m + \psi_r$ for their total.

In equilibrium, the form of the asphericity is governed by the Euler–Lagrange equation

$$\frac{\alpha}{\nu R^2 x}\frac{d}{dx}\Big(x\frac{dQ}{dx}\Big) = M(Q,x) \tag{6}$$

and the natural boundary conditions

$$\frac{\alpha}{\nu R^2}\frac{dQ}{dx}\Big|_{x=0} = \frac{\alpha}{\nu R^2}\frac{dQ}{dx}\Big|_{x=1} = 0, \tag{7}$$

with M given by

$$M(Q,x) = \frac{\mu A^2}{6\nu(1+Q)^{\frac{2}{3}}}\Big(\frac{1}{A^6} - \frac{1-Q}{1+Q}\Big) + \frac{Q}{(1+Q)^3}\Big((Q-q_*)^2\Big(1+\frac{Q(1+Q)}{Q-q_*}\Big) + \frac{\kappa A^2}{\nu R^2 x^2}\Big). \tag{8}$$

3 Numerical results

The boundary-value-problem (6)–(7) was solved numerically using the ACDC package of Cash and Wright [19]. In so doing, we set $\mu = \nu = 10^5$ J/m^3, $\kappa = \alpha = 10^{-11}$ J/m, and $R = 1$ cm. The values of μ and κ are realistic and in line with those used by Verwey, Warner and Terentjev [20] in their work on stripes in nematic elastomers. As a result of these choices, $\alpha/\nu R^2 = \kappa/\nu R^2 = 10^{-12}$. Further, for illustrative purposes, we took $q_* = -0.3$. The only parameter varied was A, the degree of cylinder distortion. We studied only the case of extension and let A range between 0.86 and 1. Figure 2 shows a sharp transition between isotropic ($q = 0$) and anisotropic ($q \neq 0$) regions along the cylinder radius, thereby indicating the presence of a disclination. For a given value of A, the value of the asphericity at the outer radius was used to determine the stability of the disclination. For an asphericity above the first inflection point of f (corresponding to $A \approx 0.97$), there is no disclination; if its value falls between the two inflection points of f, we interpret that as being an *unstable disclination*, while one beyond the second inflection point of f (corresponding to $A \approx 0.92$) is referred to as a *stable disclination*. A state connecting the energetically preferred values 0 and q_* of the asphericity is generated for $A \approx 0.90$. Following Mottram & Hogan [5], the extent of the core can also be inferred from the plot as the region where Q exhibits a rapid decrease. It is seen that, for our case, the core radius is on the order of 0.01 μm ($x = 10^{-6}$), which is of the same order as values reported by Chandrasekhar and Ranganath [21] for liquid crystalline melts.

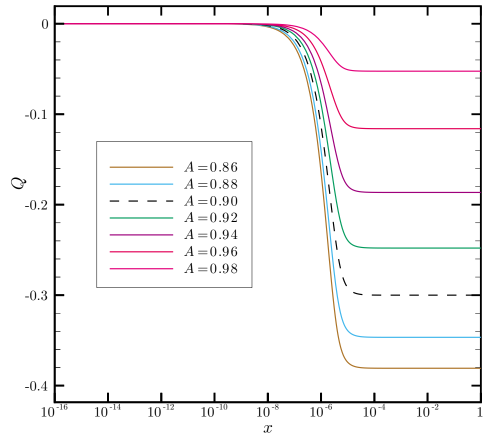

Figure 2: Plots of the asphericity Q as a function of dimensionless radial position x (note logarithmic scale) for representative values of the degree A of cylinder distortion between 0.86 and 1. Consistent with (7), note the horizontal slopes at the cylinder center ($x = 0$) and outer boundary ($x = 1$).

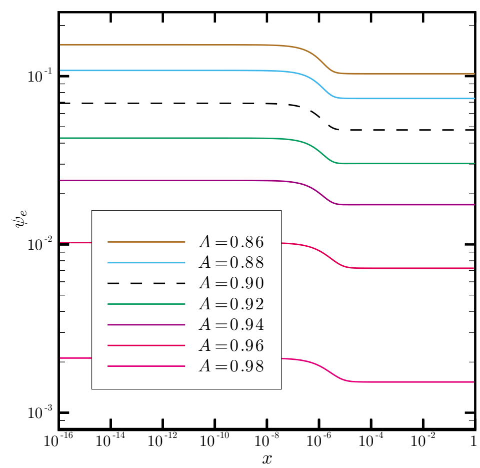

Figure 3: Plots of the rubber-elastic contribution ψ_e to the free-energy density as a function of dimensionless radial position x (note logarithmic scale) for representative values of the degree A of cylinder distortion between 0.86 and 1.

For each A, the rubber-elastic contribution ψ_e to the free-energy density is shown in Figure 3, while the difference, $\psi - \psi_e$, involving the remaining terms appears in Figure 4. In addition the free-energy subtotals $\int_0^1 \psi_e \, dx$ and $\int_0^1 (\psi - \psi_e)\, dx$ are shown in Figure 5.

From Figures 3 and 4, it is seen that the free-energy density has a markedly different behavior below and above $x \approx 10^{-6}$. This confirms our initial estimate of the core region from Figure 2. Outside of the core

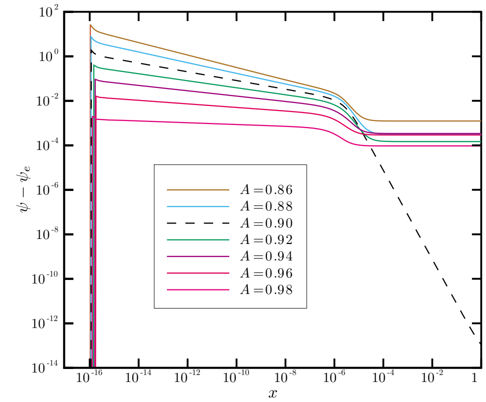

Figure 4: Plots of the portion $\psi - \psi_e$ of the free-energy density excluding the rubber-elastic contribution as a function of dimensionless radial position x (note logarithmic scale) for representative values of the degree A of cylinder distortion between 0.86 and 1.

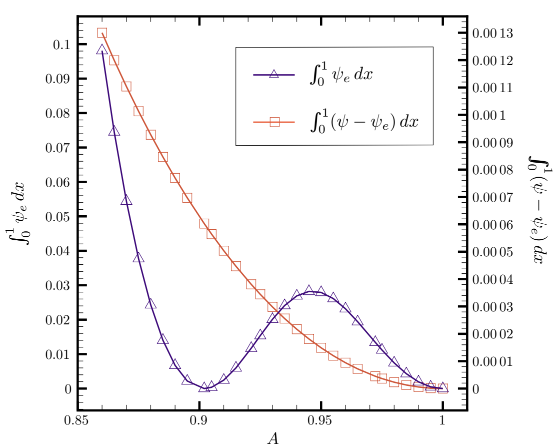

Figure 5: Plots of the portion $\int_0^1 \psi_e \, dx$ of rubber-elastic energy and of the portion $\int_0^1 (\psi - \psi_e) \, dx$ of free energy excluding the rubber-elastic contribution as a function of the degree A of cylinder distortion.

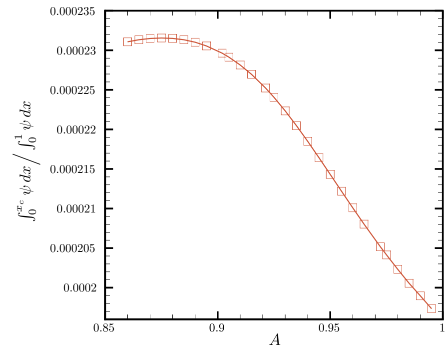

Figure 6: Plot of the percentage $\int_0^{x_c} \psi \, dx / \int_0^1 \psi \, dx$ of free energy in the core as a function of the degree A of cylinder distortion.

region, ψ_e dominates the others by two orders of magnitude. Since it is this term which describes the deformation of the bulk material as a whole, this result is not surprising.

Figure 5 indicates an expected monotonic increase with of $\int_0^1 \psi_e \, dx$ with increasing cylinder elongation (decreasing A) and also shows that the remaining contributions $\int_0^1 (\psi - \psi_e) \, dx$ to the free energy emulate the behavior of the double-well potential f. The well at $A \approx 0.90$ corresponds to the cylinder having our chosen q_* at the outer radius, while the well at $A = 1$, i.e., no stretching, is just the isotropic case of $q = 0$.

In addition, the energy of the core relative to that of the whole domain was investigated. With the extent of the core denoted as x_c and taken to be at 1.5×10^{-6} (0.015 μm), it is seen in Figure 6 that the total core energy is a vanishingly small percentage of the total energy. As mentioned before, this is because rubber elasticity persists throughout the entire domain. The remaining terms of the energy show that these are more closely tied to the formation of the core itself. In particular, for $A \approx 0.90$, the behavior within the core is interesting. From Figure 7, it is seen that the proportion $\int_0^{x_c} \psi_n \, dx \Big/ \int_0^1 \psi_n \, dx$ of nematic elastic energy contained in the core is minimized when $A \approx 0.90$—indicating that, as far as this term is concerned, a state connecting the energetically preferred values of asphericity is favored when a disclination is present. Conversely, in Figure 8, the proportion $\int_0^{x_c} (\psi_m + \psi_r) \, dx \Big/ \int_0^1 (\psi_m + \psi_r) \, dx$ of the total of the memory and regularizing terms contained in the core is maximimized ($> 70\%$) when $A \approx 0.90$. This can be explained by considering the condition of the material at this point. Here, ψ_m is minimized (practically vanishes) across the whole domain since

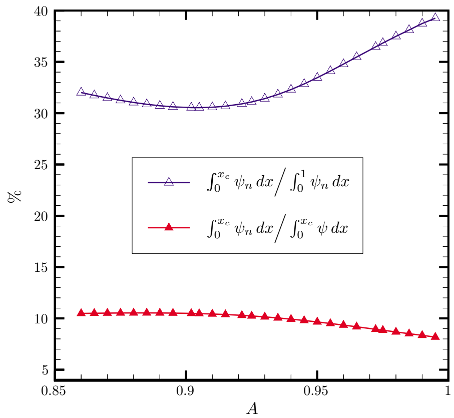

Figure 7: Plots of the percentage $\int_0^{x_c} \psi_n \, dx \Big/ \int_0^1 \psi_n \, dx$ of nematic elastic energy in the core and of the proportion $\int_0^{x_c} \psi_n \, dx \Big/ \int_0^{x_c} \psi \, dx$ of the total core energy associated with nematic elasticity for A ranging between 0.86 and 1.

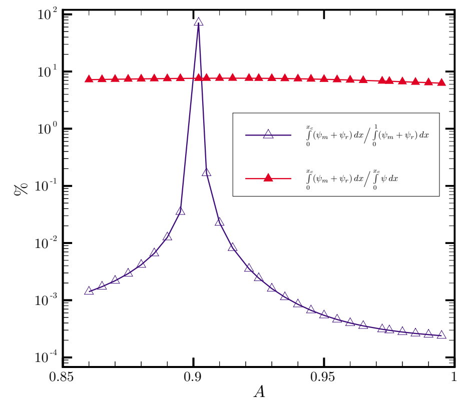

Figure 8: Plots of percentage $\int_0^{x_c} (\psi_m + \psi_r) \, dx \Big/ \int_0^1 (\psi_m + \psi_r) \, dx$ of memory and regularization energy in the core and of the proportion $\int_0^{x_c} (\psi_m + \psi_r) \, dx \Big/ \int_0^{x_c} \psi_m \, dx$ of the total core energy associated with memory and regularization for A ranging between 0.86 and 1.

all of the molecules away from the core possess an asphericity close to q_*, while the asphericity of those in the core is close to 0 (see Figure 5). However, owing to the small extent of the core and the necessity to rapidly transition from 0 to q_* (resulting in a high asphericity gradient), ψ_r is much greater in the core than in the region with asphericity q_*. Factoring in the minimization of ψ_m, it is therefore reasonable that the percentage of energy carried by ψ_m and ψ_r in the core is maximized at $A \approx 0.90$. Nevertheless, despite the variation with A of the proportions of nematic elastic energy and the sum of memory and regularizing energy within the core, the amounts of these quantities relative to total core energy show much less fluctuation (Figures 7 and 8). This implies that the percentages of those terms comprising the core energy remain relatively constant regardless of A.

4 Discussion

A rigorous theoretical basis for the development of disclinations in nematic elastomers has been introduced. Numerical results for the test case of the isochoric extension of a nematic-elastomeric cylindrical specimen were presented. They predict, for the first time, that a disclination of strength +1 will appear at the cylinder's center for a sufficiently large extension ratio. Perhaps not surprisingly, the core radius was found to be on par with that found for traditional liquid crytalline melts. While our predictions are confined to nematic elastomers which have been specially prepared, we speculate that disclinations may occur under other circumstances. We look forward to experiments designed to test our conjectures.

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938	Bagchi, P., and S. Balachandar	Linearly varying ambient flow past a sphere at finite Reynolds number—Part 1: Wake structure and forces in steady straining flow	Apr. 2000
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957	Chaïeb, S., and J. Sutin	Growth of myelin figures made of water soluble surfactant—Proceedings of the 1st Annual International IEEE–EMBS Conference on Microtechnologies in Medicine and Biology (October 2000, Lyon, France), 345–348	Oct. 2000
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960	Zhang, S., K. J. Hsia, and A. J. Pearlstein	Potential flow model of cavitation-induced interfacial fracture in a confined ductile layer—*Journal of the Mechanics and Physics of Solids* (submitted)	Nov. 2000
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